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PRELIMINARY REPORTS OF PROGRESS - LABORATORY STUDIES, WATER DECONTAMINATION. III. STUDIES ON Ce144, Y91, AND I131

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MEMORANDUM REPORT

To: Roy J. Morton

May 3, 1951

From: Conrad P. Straub, T. W. Brockett, Jr., and Robert Stepp

Subject: PRELIMINARY REPORTS OF PROGRESS - LABORATORY STUDIES
WATER DECONTAMINATION
III. STUDIES ON Ce^{144}, Y^{91}, and I^{131}

Radiochemical analyses of the effluent obtained from the alum treatment of a simulated bomb blast mixture in the experimental water treatment plant[1] showed that radioelements were removed in varying amounts. These data also indicated higher and lower removals than those that had been reported earlier when only single radioisotopes were subjected to similar treatment processes. For example, I^{131} did not show any measurable removal as a result of alum coagulation; yet in the studies referred to above[1], removals of about 40 per cent were indicated for sedimentation and filtration accounted for an additional removal of 53 per cent for a total removal of about 70 per cent. As a result, a series of jar test studies was planned in which single isotopes and mixtures of isotopes would be subjected to similar treatment processes to obtain data on removals. Such studies would show whether certain radioisotopes would be more easily removed when combined with other isotopes or whether such combinations would interfere with removals of the specific isotopes.

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PROCEDURE

In the studies performed, the procedure was quite similar in that to each beaker in the series either a single or a combination of the three isotopes was added to 500 ml of tap water solution containing

(1) Conrad P. Straub, T. W. Brockett, Jr., and Robert Stepp. Preliminary Reports of Progress. Experimental Water Treatment Plant. IV. Mixed Fission Products. To Roy J. Morton, May 1, 1951.

100 ppm of clay (turbidity). In the first and second series, the effect of mixing (rapid and slow) was investigated; in the third, the effect of pH on a mixture of the three radioisotopes, Ce, Y, and I was studied; and in the fourth, the effect of variable pH on the removals of Ce, Y, and I alone was studied.

Samples were collected of the mixed and settled supernatants, dried under infra-red lamps, and counted by means of a G-M end-window counter.

The actual procedure used in each series will be described. In the first series, to 500 ml of tap water containing 100 ppm of clay turbidity were added amounts of the individual radioisotopes or combinations thereof, and the beakers were subjected to 5 minutes of rapid and 25 minutes of slow mixing, which was followed by one-hour settling. In the second series, the samples received 30 minutes of rapid mixing and were settled for 17-1/2 hours. In the third series, a mixture of Ce, Y, and I were added to a series of beakers containing 500 ml of tap water plus 100 ppm of clay. The pH in each beaker varied, ranging from 2.0 to 11.0 as follows: 2.0, 2.8, 4.0, 5.1, 6.1, 7.0, 8.1, 9.1, 10.0 and 11.0. In series four, each radioisotope was added to a series of beakers having a pH of 3.05, 4.00, 4.98, 6.00, 7.00, 7.95, 9.00, 10.00, 11.00 and 12.00, respectively. The samples were rapid-mixed for 30 minutes and allowed to settle for 4 hours.

RESULTS

Series 1 and 2. Effect of Mixing

The results shown in Table I indicate that slow mixing was not conducive to maximum removals. Table II, which reports the results obtained with 30 minutes of rapid mixing and prolonged settling (17-1/2

hours), shows much greater removals for Ce and Y.

Series 3 and 4. Effect of pH

To check the effect of pH on the removal of activity by 100 ppm of turbidity (clay) from a mixture of radioisotopes containing Ce^{144}, I^{131}, and Y^{91}, a series of beakers having variable pH's, as shown in Table III, was set up. As can be seen from the data presented, pH exerted a tremendous effect on removals. The results are plotted in Figure 1.

The per cent activity removed from the composite mixture is computed as follows:

Ce^{144} = 451 cts/min/ml

I^{131} = 346

Y^{91} = 596

1393 cts/min/ml in the initial mixture

Per cent removal = $\frac{1393 - 1201}{1393} \times 100 = 14$

Additional studies were carried out using individual radioisotope solutions of Ce^{144}, I^{131}, and Y^{91} to determine the effect of pH on removals. The results obtained are given in Tables IV, V, and VI, and in Figures 2, 3, and 4.

DISCUSSION

The first studies show conclusively the beneficial effect of rapid mix and long-time settling on removal. With the coarse clay used in these studies, rapid mixing is necessary to keep the clay in suspension. Slow mixing permits the clay to settle out, reducing its contact with the radioactive environment. In all probability, if finer clays were used it would have been possible to utilize slow mixing.

The values reported in Table II show that both Ce^{144} and Y^{91} will be adsorbed on clays, the removals being 93 and 94 per cent, respectively. Combinations of Ce and Y show removals of 94 per cent when

100 ppm clay was used and 99 per cent with 200 ppm clay. In the combination Ce and I, if it is assumed that no I was removed, it will be found that a removal of 99 per cent of the Ce was obtained. Similar computations for the I and Y combination showed a removal of 98 per cent for the Y. The Ce, I, and Y combinations, again assuming that no I was removed, gave total Ce and Y removals of 99.9 per cent, 98.3 per cent, and 96.4 per cent for 100, 200, and 300 ppm of clay, respectively.

In the several series in which removals of Ce, I, and Y were checked individually and in combination at variable pH levels, it was found that considerably higher removals were obtained in the alkaline region. Removals of 70 per cent or more were obtained at a pH of 7 or above. The study of the mixture of Ce, I, and Y is of interest, since in jars 9 and 10 it was found that if values for the removal of Ce and Y were assigned to the removal values in agreement with results in the series involving mixing only, that removal of the I was obtained along with the Ce and Y. The calculations are given below and the values used are taken from Table III.

If the $Ce^{144} + Y^{91}$ is completely removed it would account for a reduction of

$$\frac{451 + 596}{1393} \times 100 = \frac{1047}{1393} \times 100 = 75\%$$

Since the removals indicated for jars 9 and 10 are greater than this, some of the I^{131} apparently has been removed as well. If we continue to assume complete removal of the Ce and Y, then the amounts of I removed in jars 9 and 10 would be

$$\frac{346 - 268}{346} \times 100 = \frac{78}{346} \times 100 = 23\% \text{ removal for } I^{131} \text{ from jar 9}$$

From jar 10 removal of I^{131} will be

$$\frac{346 - 316}{346} \times 100 = 9\%$$

If we assume removals of 94 per cent for the $Ce^{144} + Y^{91}$ (when 100 ppm of clay are added) as indicated in Table II the amount of I^{131} removed from jar 9 will be 1393 - 346 = 1047 and 6% of Ce and Y remain then

$$\frac{346 - 268 + (1047 \times 0.06)}{346} \times 100 = \frac{140.8}{346} \times 100 = 40.7\%$$

These calculations indicate the possible formation of Ce and/or Y-iodine compounds. A cursory check of the literature reveals the existence of such compounds in the form of $Ce(IO_3)_4$, $CeI_3 \cdot 9H_2O$, and YI_3. If these compounds are formed and adsorbed on clay particles, then the removals for the I would appear reasonable. If these compounds are not formed, then no explanation (with the possible exception of self-absorption) can be offered at present for the indicated removal of the I, since as shown in the case of I above, there is no adsorption of I on clay in this range. The only removals indicated are in the acid range and these are not removals but probably only the loss of free I_2 by volatization in an acid medium. Both Ce and Y singly show higher removals at increased pH values and we find that, in general, higher removals occur at higher pH values.

CONCLUSIONS

These studies show that (1) for adsorption studies involving the clay used it will be necessary to utilize rapid mix (and possibly long-time settling, although this is not believed necessary) if effective removals are desired; (2) for Ce, I, and Y singly or in combination

the Ce and Y may be effectively removed, whereas no removal of the I is effected; (3) at high pH values high removals of Ce and Y can be obtained; (4) iodine is not influenced in any way by pH (no removal at any pH value); and (5) when combined with Ce and Y, apparent removals of I are shown at high pH values, indicating the possible formation of Ce- and Y-iodine compounds.

CPStraub/vcm

7/12/51

Table I

Removal of Ce, I, and Y by Turbidity

Rapid Mix 5 Minutes - Slow Mix 25 Minutes

Radioisotope	Activity cts/min/ml* Initial	Final	Per Cent Removal
Ce^{144}	298	223	25
I^{131}	437	419	0
Y^{91}	346	212	39
Ce + I	484	438	10
I + Y	375	317	16
Ce + Y	465	316	32
Ce + I + Y	857	652	24

*Uncorrected for geometry

Table II

Removal of Ce, I, and Y by Turbidity

Rapid Mix - 30 Minutes

Radioisotope	ppm Clay	Activity cts/min/ml* Initial	Activity cts/min/ml* Final	Per Cent Reduction	pH After Addition of Radioisotopes**
Ce^{144}	100	572	41	93	7.6
I^{131}	100	435	453	--	7.6
Y^{91}	100	299	17	94	7.5
$Ce^{144} + I^{131}$	100	994	411	59	7.5
$I^{131} + Y^{91}$	100	693	415	40	7.4
$Ce^{144} + Y^{91}$	100	856	54	94	7.6
$Ce^{144} + Y^{91}$	200	913	13	99	7.6
$Ce^{144} + I^{131} + Y^{91}$	100	1263	409	68	7.7
$Ce^{144} + I^{131} + Y^{91}$	200	1279	423	67	7.5
$Ce^{144} + I^{131} + Y^{91}$	300	1219	437	64	7.6

* Uncorrected for geometry

** ph - Tap water - 7.0, Tap water + clay 7.4

Table III

Removal of Activity by Clay from Solutions of Variable pH*

Beaker	Before Clay Addition	After Clay Addition	Activity cts/min/ml** Initial	Final	% Removal
Control			1393***		
1	2.0	2.0		1201	14
2	2.8	2.84		1017	27
3	4.0	4.45		955	32
4	5.1	6.85		570	59
5	6.1	7.85		371	73
6	7.0	8.20		421	70
7	8.1	8.30		419	70
8	9.1	8.40		456	67
9	10.0	9.10		268	81
10	11.0	10.00		316	77
Ce^{144}			451		
I^{131}			346		
Y^{91}			596		

*Mixture contains Ce^{144}, I^{131}, and Y^{91}

**Uncorrected for geometry

***Calculated from Ce^{144}, I^{131}, and Y^{91} data given

Table IV

Removal of Ce^{144} by 100 ppm Clay at Variable pH Values

Beaker	pH Initial	pH Final	Activity* - cts/min/ml Initial	Activity* - cts/min/ml Final 4 hrs. Settling	Activity* - cts/min/ml Final 24 hrs. Settling	% Reduction 4 hrs. Settling	% Reduction 24 hrs. Settling
1	3.05	3.1	492	434	424	12	15
2	4.00	3.5	451	341	317	24	30
3	4.98	8.0	526	131	42	75	98
4	6.00	8.45	445	117	38	74	92
5	7.00	8.4	420	175	39	58	91
6	7.95	8.15	420	149	59	65	86
7	9.00	8.48	418	132	51	68	88
8	10.00	9.52	439	61	19	86	96
9	11.00	10.35	437	8	4	98	99
10	12.00	11.85	429	24	35	94	92

*Uncorrected.

Table V

Removal of I^{131} by 100 ppm Clay at Variable pH

Beaker	pH			Activity* - cts/min/ml			% Reduction	
	Initial	Final 4 hrs. Settling	Final 24 hrs. Settling	Initial	Final 4 hrs. Settling	Final 24 hrs. Settling	4 hrs** Settling	24 hrs** Settling
1	3.05	3.3	3.28	952	823	736	6	8
2	4.15	4.45	4.09	888	756	700	7	6
3	5.6	6.7	6.82	238	188	184	1	1
4	6.6	7.6	7.78	205	210	225	-	-
5	7.5	8.1	8.82	231	221	200	-	-
6	8.0	8.2	8.38	268	278	241	-	-
7	8.8	8.45	8.46	348	413	397	-	-
8	9.75	9.3	8.87	296	291	265	-	-
9	10.7	10.2	9.81	296	275	277	-	-
10	11.8	11.7	11.25	299	278	276	-	-

* Uncorrected for geometry.
** % removal corrected for loss due to decay.

Table VI

Removal of Y^{91} by 100 ppm Clay at Variable pH

Beakers	pH			Activity - cts/min/ml			% Reduction	
	Initial	Final 4 hrs. Settling	Final 24 hrs. Settling	Initial	Final 4 hrs. Settling	Final 24 hrs. Settling	4 hrs. Settling	24 hrs. Settling
1	3.1	3.05	3.03	677	581	592	14	13
2	4.12	6.18	6.42	646	321	196	50	70
3	5.65	6.83	6.77	679	192	97	72	86
4	6.7	7.72	7.59	672	164	37	76	94
5	7.4	8.16	8.05	650	172	62	74	90
6	7.9	8.33	8.28	623	168	66	73	89
7	8.8	8.43	8.32	638	215	65	66	90
8	9.72	9.38	8.92	636	121	46	81	93
9	10.7	10.24	9.83	620	47	18	92	97
10	11.9	11.77	11.32	616	89	116	86	81

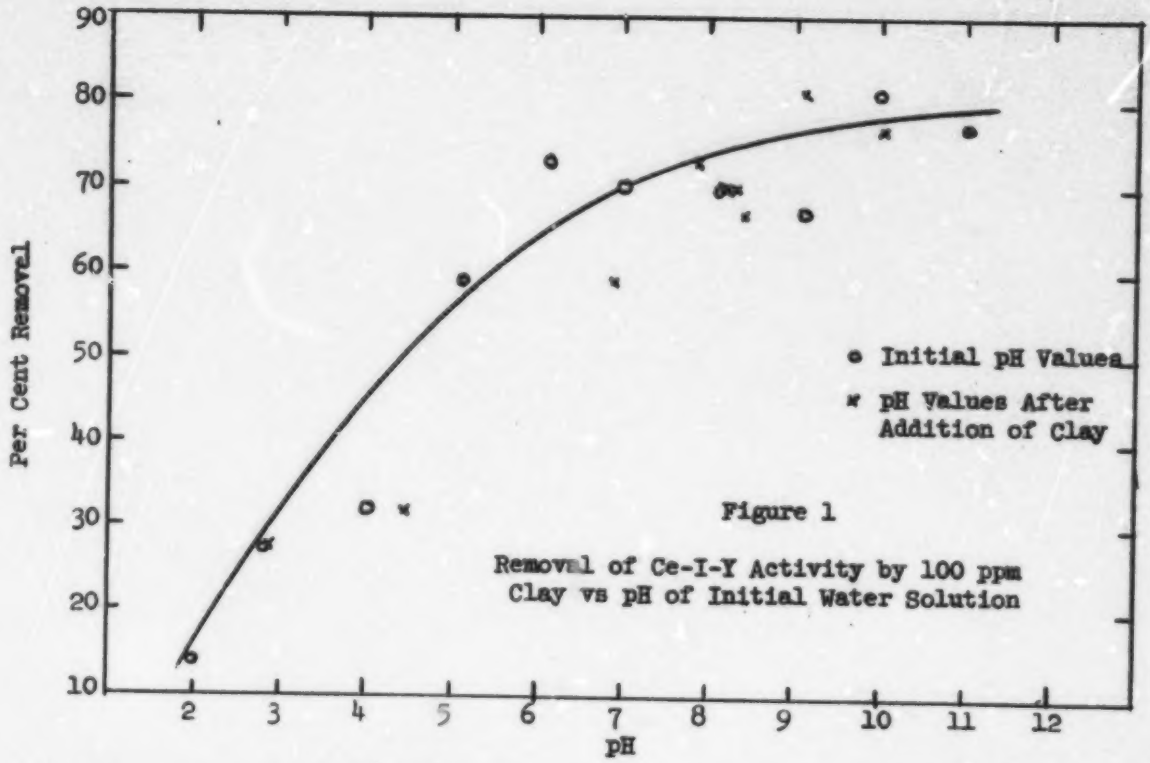

Figure 1

Removal of Ce-I-Y Activity by 100 ppm Clay vs pH of Initial Water Solution

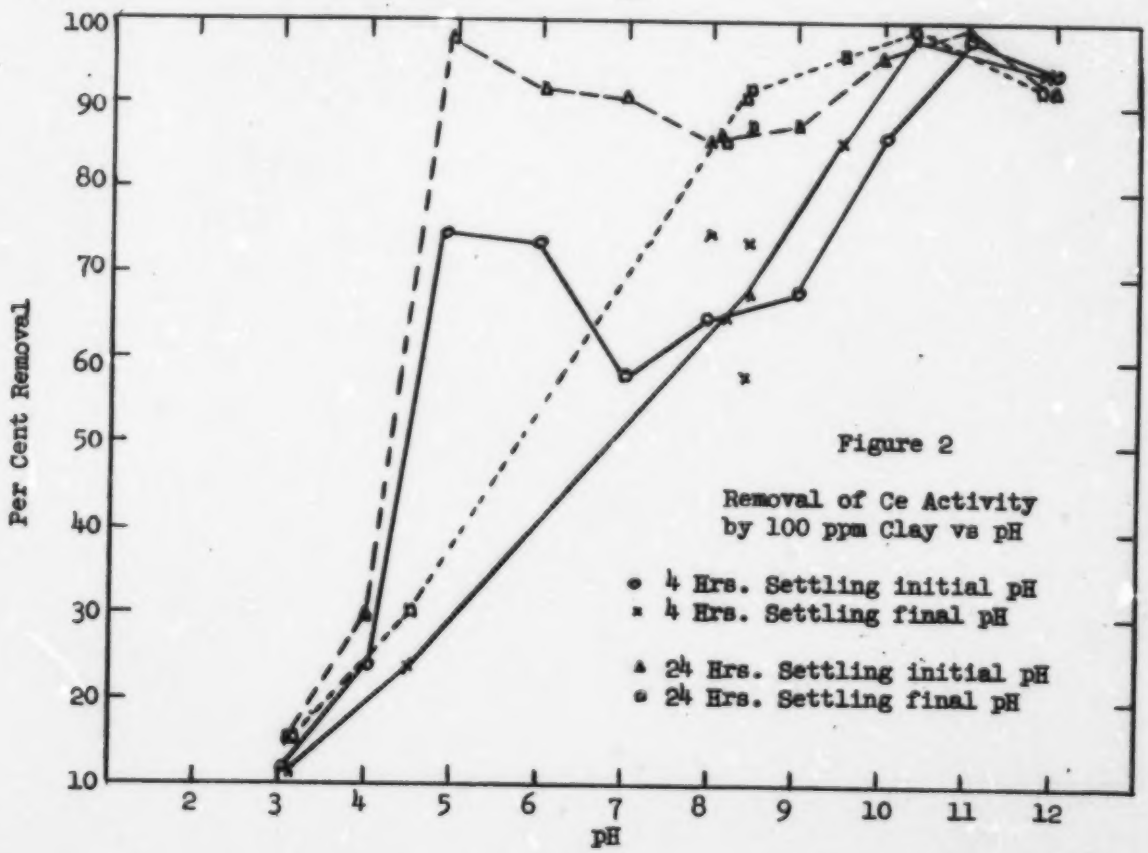

Figure 2

Removal of Ce Activity by 100 ppm Clay vs pH

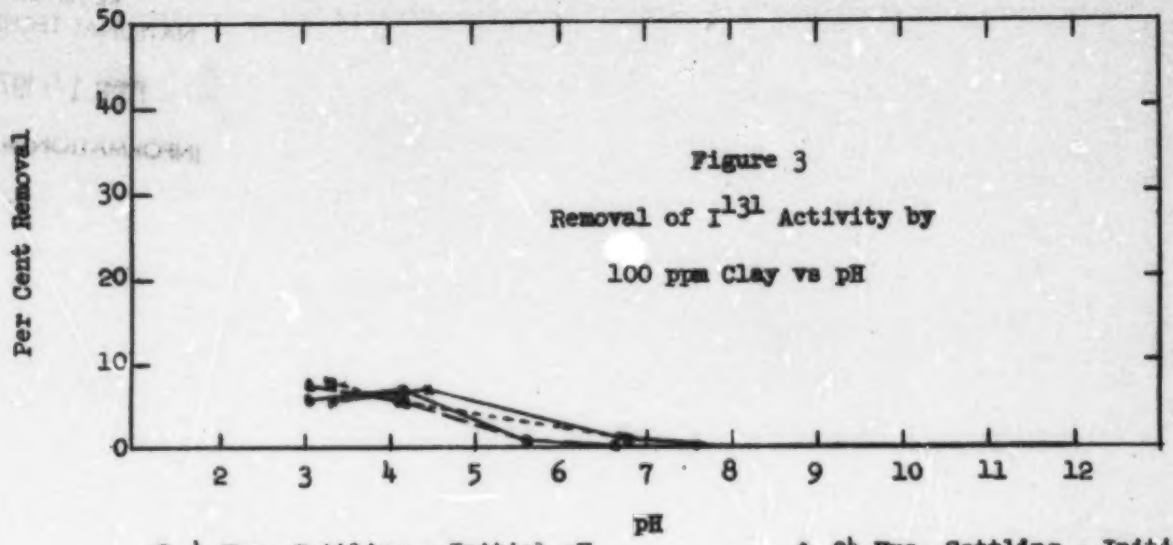

Figure 3

Removal of I^{131} Activity by

100 ppm Clay vs pH

o 4 Hrs. Settling - Initial pH

x 4 Hrs. Settling - Final pH

▲ 24 Hrs. Settling - Initial pH

□ 24 Hrs. Settling - Final pH

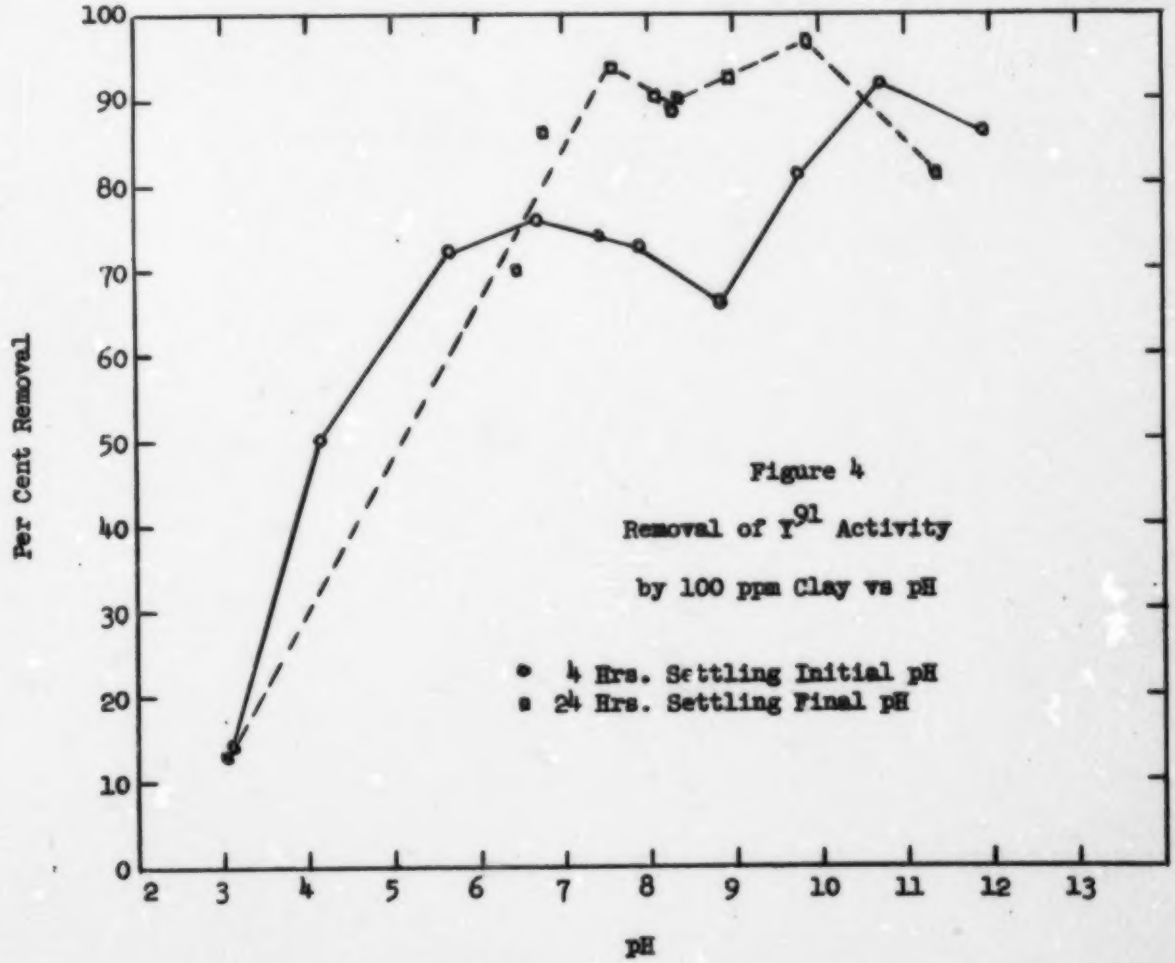

Figure 4

Removal of Y^{91} Activity

by 100 ppm Clay vs pH

o 4 Hrs. Settling Initial pH

□ 24 Hrs. Settling Final pH